

Electric Nusselt number characterization of electroconvection in nematic liquid crystals

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Abstract

We develop a characterization method of electroconvection structures in a planar nematic liquid crystal layer by a study of the electric current transport. Because the applied potential difference has a sinusoidal time dependence, we define two electric Nusselt numbers corresponding to the in-phase and out-of-phase components of the current. These Nusselt numbers are predicted theoretically using a weakly nonlinear analysis of the standard model. Our measurements of the electric current confirm that both numbers vary linearly with the distance from onset until the occurrence of secondary instabilities; these instabilities also have a distinct Nusselt number signature. A systematic comparison between our theoretical and experimental results, using no adjusted parameters, demonstrates reasonable agreement. This represents a quantitative test of the standard model completely independent from tradi-

tional, optical techniques of studying electroconvection.

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Although spontaneous pattern formation within structureless environments pervades nature [1], a comprehensive understanding of this complex behavior remains elusive. Therefore well-controlled experimental systems exhibiting pattern formation are extensively studied; among these, thermoconvection of a layer of fluid heated from below [2] and electroconvection of a nematic liquid crystal layer [3,4] are particularly interesting since they allow very large aspect ratio geometries. In both of these systems, convection structures form spontaneously when the applied stress, i.e. the gradient of either the temperature or the electric potential, exceeds a critical value. These inherently non-equilibrium structures can only persist when there is an energy source to overcome the dissipation associated with the flow. Therefore, energy transport studies represent a particularly valuable technique for elucidating the essential nature of the instabilities that lead to these patterns. For example, the first accurate determination of the stress necessary to induce thermoconvection was made by measuring the heating power required to sustain a desired temperature difference across a thin layer of water [5]. This power is customarily expressed as the Nusselt number, defined as the heat flow across a fluid layer relative to the heat flow required in the absence of fluid flow. Nusselt number measurements remain a method of choice for studies not only of the structured states that occur during thermoconvection when the stress is only slightly above its critical value [6] but also of the turbulent flow that occurs when the stress is enormous [7]. By contrast the electroconvection of a planar nematic liquid crystal layer, which represents a similar but fully *anisotropic* model pattern forming system, has previously been studied only with qualitative or semi-quantitative optical techniques. Reports of energy flow measurement during electroconvection are rare [8], and no theoretical studies of the energy transport exist for this system. The aim of this Letter is to fill this gap.

Electroconvection is obtained when an a.c. electric potential, $\sqrt{2}V \cos(\omega t)$, is applied to two horizontal ($\perp \hat{\mathbf{z}}$) electrodes separated by distance d confining a nematic liquid crystal. Here we focus on the *planar* anchoring case where the director field \mathbf{n} is fixed to $\hat{\mathbf{x}}$ at the confining electrode plates. The instability relies on a coupling between \mathbf{n} , the velocity field, \mathbf{v} and the induced charge density, ρ_e or equivalently the induced electric potential ϕ , such

that the full electric field reads $\mathbf{E} = \sqrt{2}V/d [\cos(\omega t)\hat{\mathbf{z}} - d\nabla\phi]$. At moderate frequencies ω , when V exceeds a critical value V_c , the instability sets in the form of normal *conduction* rolls of wavevector $\mathbf{q} = q\hat{\mathbf{x}}$; at large frequencies *dielectric* rolls are observed [4] but we do not consider this regime in this work. These phenomena are well explained via the standard model (SM) for electroconvection [3,4] where the charge conduction in the liquid crystal is assumed Ohmic. In addition to linear properties (values of V_c and q as a function of ω), the SM explains several secondary instabilities that are experimentally observed, such as the transitions to zig-zag rolls, stationary and oscillatory bimodal patterns and abnormal rolls [9]. The one phenomenon which the SM has been unable to predict is the traveling roll state. For this, a new approach, the weak electrolyte model, was developed, in which the electrical conductivity is assumed to be due to two species of dissociated ions having different mobilities [10]. With this model a semi-quantitative agreement with experimental results on traveling rolls was demonstrated [10] but fitting parameters were necessary. Because traveling rolls are not encountered at low frequencies where we performed our experiments, and because the SM is much less complicated, we did not use the weak electrolyte model.

The total current I through the nematic cell enclosed by the horizontal electrodes of area S can be calculated as the circulation of the magnetic induction \mathbf{H} . From the Maxwell-Ampère equation, $\nabla \times \mathbf{H} = \mathbf{j} + \partial_t \mathbf{D}$, I is sum of the conduction and displacement currents:

$$I = \iint_S (j_z + \partial_t D_z) dx dy \quad (1)$$

where, within the SM, $\mathbf{j} = \sigma_\perp \mathbf{E} + \sigma_a(\mathbf{n} \cdot \mathbf{E})\mathbf{n} + \rho_e \mathbf{v}$; $\mathbf{D} = \epsilon_\perp \mathbf{E} + \epsilon_a(\mathbf{n} \cdot \mathbf{E})\mathbf{n}$; σ_\perp (σ_\parallel) are the conductivities perpendicular (parallel) to \mathbf{n} , and, ϵ_\perp (ϵ_\parallel) are the dielectric permittivities perpendicular (parallel) to \mathbf{n} , $\sigma_a = \sigma_\parallel - \sigma_\perp$ and $\epsilon_a = \epsilon_\parallel - \epsilon_\perp$ are the corresponding anisotropies. Note that the surface integral in eq. (1) does not depend on the z -value ($-d/2 \leq z \leq d/2$) chosen, because of the Maxwell-Ampère equation. In the quiescent (no convection) state $\mathbf{n} = \hat{\mathbf{x}}$, $\mathbf{v} = \mathbf{0}$ and $\phi = 0$, therefore

$$I = I^0 = I_r^0 \cos(\omega t) - I_i^0 \sin(\omega t) = \frac{\sqrt{2}VS}{d} [\sigma_\perp \cos(\omega t) - \epsilon_\perp \omega \sin(\omega t)] . \quad (2)$$

In the convecting state all fields \mathbf{n}, \mathbf{v} and ϕ are modified, as is I . Within the SM, for homogeneous stationary roll solutions

$$I = I_r \cos(\omega t) - I_i \sin(\omega t) + \text{higher temporal harmonics} \quad (3)$$

where the amplitudes of the higher temporal harmonics are expected to be much smaller than I_r and I_i , at least at intermediate frequencies $\omega \simeq 1/\tau_0$ where $\tau_0 = \epsilon_\perp/\sigma_\perp$ is the charge-diffusion time. We define the real and imaginary reduced Nusselt numbers, \mathcal{N}_r and \mathcal{N}_i , as $I_r/I_r^0 - 1$ and $I_i/I_i^0 - 1$, respectively. Thus $\mathcal{N}_r = \mathcal{N}_i = 0$ in the quiescent state, while in the convecting state \mathcal{N}_r measures the excess energy dissipation due to convection of the nematic liquid crystal, that is the time average $\langle \sqrt{2}V \cos(\omega t) I(t) \rangle_t = (1 + \mathcal{N}_r) V^2 \sigma_\perp S/d$. In heuristic terms the effective resistance of the nematic layer is changed by convection from $R_0 = d/(\sigma_\perp S)$ to $R = R_0/(1 + \mathcal{N}_r)$; equivalently the imaginary Nusselt number measures the change in the effective capacitance of the nematic layer, $C_0 = \epsilon_\perp S/d$ in the quiescent state, $C = C_0(1 + \mathcal{N}_i)$ in the convecting state. When the reduced distance from onset $\epsilon \equiv V^2/V_c^2 - 1$ is small, the electric Nusselt numbers can be calculated for homogeneous rolls using weakly nonlinear methods. Assuming that the leading convection amplitude, A , associated with the linear roll mode, remains small, a systematic expansion in powers of A is performed. After adiabatic elimination of the slave modes and calculation of the resonant saturating cubic terms approximate roll solutions are obtained together with the relation $A(\epsilon) = a_0 \sqrt{\epsilon}$. The current can then be calculated from eq. (1). For symmetry reasons the first contribution from the convection modes comes at order A^2 , and therefore one expects $\mathcal{N}_r \sim A^2 \sim \epsilon$, $\mathcal{N}_i \sim A^2 \sim \epsilon$ in the weakly nonlinear regime. That is, Nusselt numbers allow a direct measurement of the convection amplitude A , and therefore a test of the supercritical law $A(\epsilon) = a_0 \sqrt{\epsilon}$. In order to make this clear and to obtain *approximate* analytic formulae, one can first use the quasi-unidimensional approximation, where all fields are considered at the middle of the layer ($z = 0$) and only their x -dependence is kept. The linear normal roll mode then assumes the form $n_z = -AN_z \sin(qx)$, $v_z = A/(q\tau_0)V_z \cos(qx)$, $\phi = A/(qd)[\Phi_c \cos(\omega t) + \Phi_s \sin(\omega t)] \cos(qx)$ where, as in the rest of our

theoretical calculations, we only keep the lowest nontrivial time-mode for each field. We also choose as a normalization condition $N_z = 1$; then V_z , Φ_c and Φ_s are calculated at fixed frequency by solving the linear neutral eigenproblem. With $\rho_e = \nabla \cdot \mathbf{D}$, $j_z + \partial_t D_z$ can be easily calculated at $z = 0$. Keeping only the horizontally homogeneous terms because of the surface integral in eq. (1), one obtains to lowest order in A

$$\mathcal{N}_r = \frac{A^2}{2} [\sigma'_a N_z (N_z - \Phi_c) + \epsilon'_\parallel \Phi_c V_z - \epsilon'_a N_z (V_z + \omega \tau_0 \Phi_s)] \quad (4)$$

$$\mathcal{N}_i = \frac{A^2}{2} \left[\epsilon'_a N_z (N_z - \Phi_c) + \frac{\Phi_s}{\omega \tau_0} (\sigma'_a N_z - \epsilon'_\parallel V_z) \right] \quad (5)$$

with $\sigma'_a = \sigma_a / \sigma_\perp$, $\epsilon'_a = \epsilon_a / \epsilon_\perp$ and $\epsilon'_\parallel = \epsilon_\parallel / \epsilon_\perp$. For standard nematic materials with large positive σ'_a (see e.g. eq. (6)), the leading term in \mathcal{N}_r eq. (4) is the anisotropic conduction term in $\sigma'_a N_z^2$, which imposes a positive value of \mathcal{N}_r . One thus expects that the tilt of the director out of the plane in roll structures will enhance the electrical conduction of the layer and finally the in-phase current. Concerning \mathcal{N}_i eq. (5) it should be noted that Φ_s / ω tends to a finite positive value when $\omega \rightarrow 0$. Two terms of eq. (5) control the sign of \mathcal{N}_i . The first term in $\epsilon'_a N_z^2$ reveals a diminution of the effective capacitance of the cell due to the director tilt, since the dielectric anisotropy ϵ'_a of the nematic materials used in electroconvection is usually negative. The other important contribution is the positive term in $\sigma'_a N_z \Phi_s / \omega$, which expresses that the potential modulation induced by the convection creates by coupling with the director tilt an out-of-phase current $I_i > 0$ (see eq. (3)). Since Φ_s / ω decreases with ω , this positive term can compensate the negative term in $\epsilon'_a N_z^2$ at low frequencies only, and for nematics with large σ'_a . A *numerical* calculation, using a standard Galerkin technique to expand the z -dependence of all fields in test functions, can provide a more accurate evaluation of \mathcal{N}_r and \mathcal{N}_i . For this purpose we have modified the code developed in [9] to use Tchebyshev polynomials as the test functions in order to accelerate the convergence (typically 4 z -modes were sufficient), and inserted a procedure to calculate the Nusselt numbers. For convenience the current eq. (1) is evaluated at the lower plate $z = -d/2$ where, because of the boundary conditions, $j_z + \partial_t D_z$ reduces to $(\sigma_\perp + \epsilon_\perp \partial_t) E_z$. Thus, since the convection induced potential is even under $z \mapsto -z$ at linear order, but odd

at quadratic order, one sees that the leading contribution to I comes from the potential part of the homogeneous quadratic slave mode noted $A^2V_2(\mathbf{q}, -\mathbf{q})$ in eq. (27) of [9]. Of course the saturation at cubic order needs also to be calculated in order to provide the law $A = a_0\sqrt{\epsilon}$. We will return to the numerical results (Fig. 3), which confirm the trends found from the analytic formulae eqs. (4), (5), after presenting our experimental results.

We use the “classical” arrangement [11] based on a pre-fabricated liquid crystal cell [12]. The glass plates have no spacers, glue, etc. within the active area; they are separated by $d = 23.4 \pm 0.5\mu\text{m}$. With air only between the plates, we measure, using a auto-balancing 1kHz bridge, the capacitance of the cell in order to determine accurately (within 8 ppm) the ratio S/d (nominally $S = 5\text{mm} \times 5\text{mm}$). After this measurement the nematic liquid crystal methoxy-benzylidene butyl-aniline (MBBA) [13], doped with 0.0005% tetrabutyl ammonium bromide, is introduced between the transparent conducting electrodes. The filled cell is placed in a temperature controlled housing, and then introduced between the pole faces of a large electromagnet. As the nematic liquid crystal undergoes the magnetically induced splay Frederiks transition, the capacitance and conductance of the cell are monitored. From these measurements we obtain both electric conductivities and both dielectric constants [14]. Hence, we measure *in situ* all the electrical transport properties of the specific nematic liquid crystal used. For the experiments reported here, all at 28°C, we find

$$\begin{aligned}\sigma_{\perp} &= (8.5 \pm 0.8) 10^{-8}(\Omega\text{m})^{-1}, \quad \sigma'_a = \sigma_a/\sigma_{\perp} = 0.35 \pm 0.04, \\ \epsilon_{\perp} &= (4.65 \pm 0.03)\epsilon_0, \quad \epsilon'_a = \epsilon_a/\epsilon_{\perp} = -0.080 \pm 0.001,\end{aligned}\tag{6}$$

with ϵ_0 the vacuum dielectric permittivity. After these measurements, the nematic cell is transferred to the stage of a polarizing microscope so that shadowgraph [15] images can be obtained concomitantly with the electric current measurements. A function generator is used to produce a sinusoidal voltage signal which is in turn amplified, and applied to the cell. The path to ground for the current traversing the cell is through a current-to-voltage converter. The output signal from this converter is measured by a lock-in amplifier, whose reference signal is supplied by the original function generator. Before any measurements are

taken, the nematic cell is replaced by a purely resistive load and the phase setting on the lock-in is adjusted to zero the out-of-phase current component. The nematic cell is then re-inserted. Then, at a selected frequency V is raised in small steps. At each step, after waiting several seconds, I_r and I_i are recorded [16]. This proceeds until a maximum desired V (well above the threshold value V_c) is reached. Then, the process is reversed, and the currents recorded as V decreased. The difference in current for increasing vs decreasing V is less than 2%. When V is raised above V_c , the electric current traversing the liquid crystal measurably deviates from its value in the quiescent state, I^0 . In order to determine V_c from either the in-phase or out-of-phase current data, we first determine a baseline for I_r^0 (I_i^0) by fitting a straight line to I_r (I_i) vs V for V much smaller than V_c ; see Fig. 1. These values of I_r^0/V and I_i^0/V provide independent measurement of σ_\perp and ϵ_\perp (see eq. (2)) that agree within 5% with the direct measurement of these parameters using the Frederiks transition. The Nusselt numbers as functions of V are then calculated by subtracting unity from the ratios I_r/I_r^0 and I_i/I_i^0 . By fitting another straight line to \mathcal{N}_r (\mathcal{N}_i) in the region where it deviates from zero, we define V_c as where this line crosses zero (see the insets in Figs. 1 and 2). For one ramp of V , the three values of V_c determined from the Nusselt numbers and the traditional shadowgraph technique agree with each other within 0.01%.

Our apparatus did not reach sufficiently large V to measure the crossover to the dielectric regime (see e.g. [4]); we therefore estimated a characteristic “cutoff” frequency ω_c by fitting $V_c(\omega)$ to the function $A/(\omega - \omega_c)$. We found typically $\omega_c/(2\pi) = 645\text{Hz}$, but during the course of taking the measurements (2-3 months) this quantity varied by $\pm 10\%$, probably in connection with variations of the electrical parameters, especially the conductivities.

In Fig. 2 we plot both the real and imaginary Nusselt numbers vs ϵ . While \mathcal{N}_r is always observed to be positive, for the data set shown \mathcal{N}_i is negative. In some cases (discussed subsequently) it becomes positive. Note also that \mathcal{N}_r is at least ten times larger in magnitude than \mathcal{N}_i .

Close to threshold, i.e. for $0 \leq \epsilon \lesssim 0.1$, both Nusselt numbers are proportional to ϵ as shown in the inset for the real Nusselt number: this confirms the supercritical law

$A \sim \sqrt{\epsilon}$. The variations of the corresponding slopes \mathcal{N}_r/ϵ and \mathcal{N}_i/ϵ vs $\omega\tau_0$ are given on Fig. 3, which represents the results of several ramps in ϵ at each frequency. For $\epsilon \gtrsim 0.1$ the curves deviate from straight lines; the “knees” in the curves indicate clearly the onset of secondary instabilities. Specifically, the two arrows shown on Fig. 2 correspond to the onset potential differences for the zig-zag instability [17] and the spontaneous generation of dislocations associated with the so-called “defect chaos” [18].

To compare our experimental results with the SM calculations, we used the elastic constants and the viscosities measured for MBBA at 28°C in [19,20], and the electric parameters that we determined independently (eq. (6)). Varying those parameters within the stated uncertainties, we calculate (with the fitting procedure defined above) for the “cutoff” frequency $\omega_c/(2\pi) = 730 \pm 120\text{Hz}$, in agreement with the measured value. Systematically varying the electrical parameters within the experimental error bars, we also calculate, with the weakly nonlinear numerical code introduced above, the bands of possible values of \mathcal{N}_r/ϵ and \mathcal{N}_i/ϵ . These bands are drawn in gray on Fig. 3. The extremal values turn out to be obtained by variation of only σ'_a , with the upper (lower) curves for both \mathcal{N}_r/ϵ and \mathcal{N}_i/ϵ obtained for the largest (smallest) value of σ'_a . This is consistent with the fact that the leading positive terms controlling the Nusselt numbers as seen in the approximate formulae eqs. (4), (5) are proportional to σ'_a . Note also that for large σ'_a we expect \mathcal{N}_i to be positive at low frequencies, while \mathcal{N}_i is always negative for σ'_a small. There is a good agreement between experiments and theory concerning the imaginary Nusselt number \mathcal{N}_i ; on the other hand the real Nusselt number \mathcal{N}_r decreases more abruptly in the experiments than in the theory. However the agreement obtained for \mathcal{N}_i at all frequencies and \mathcal{N}_r at small frequencies is particularly significant since (contrarily to the standard approach in nematic electroconvection where usually σ_\perp is fitted) no adjusted parameters have been used.

In conclusion, electric Nusselt number measurements are validated as a new and powerful method of characterization of electroconvection. This wholly quantitative technique stands in contrast to traditional optical methods which only become quantitative in certain limit cases. This technique affords a precise determination of the threshold voltage for the onset

of electroconvection as well as secondary instabilities. Nusselt number measurements also represent an important quantitative tool for testing competing theoretical descriptions of electroconvection. Here, with only the limitation of relying on tabulated values of some material parameters, we have shown that the standard model for electroconvection gives satisfactory predictions of the Nusselt numbers near onset. One conspicuous explanation for the remaining discrepancies may be that nematic liquid crystals are quite clearly electrolytic conductors, and thus the Ohmic conduction assumed in the standard model introduces an important approximation. Thus, it clearly is of interest to extend the calculations presented here within the so-called weak-electrolyte model [10]. Future directions of this work include also employing liquid crystal materials for which the applicability of the weak electrolyte model has been established, and experiments in the highly nonlinear, dynamical scattering regimes that occur at very large ϵ [21].

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FIGURES

FIG. 1. In-phase current I_r vs the applied voltage V at a frequency $\omega/(2\pi) = 100\text{Hz}$ i.e. $\omega\tau_0 = 0.30$. *Inset:* blowup of the neighbourhood of V_c .

FIG. 2. Real and imaginary reduced Nusselt numbers vs the distance to threshold ϵ for electroconvection of MBBA at the same frequency than Fig. 1. The arrow on the left indicates the onset of the secondary zig-zag instability and the arrow on the right indicates the onset of defect chaos. *Inset:* blowup of the $\epsilon \sim 0$ region indicating how \mathcal{N}_r initially increases linearly with ϵ . Small pre-transitional effects caused by sample imperfections are also seen.

FIG. 3. Black circles: measured ratios \mathcal{N}_r/ϵ and \mathcal{N}_i/ϵ at small ϵ vs the dimensionless frequency. The vertical error bars correspond to the imprecision in measuring \mathcal{N}/ϵ and to the dispersion between different ϵ ramps, while the horizontal error bars originate from the variation in the charge-diffusion time τ_0 . Gray bands: intervals of values of the same quantities deduced from the weakly nonlinear SM calculations, taking into account the variations of the electrical parameters of the nematic liquid crystal eq. (6). The upper (lower) theoretical curves on both graphs were calculated with the largest (smallest) value of $\sigma'_a = \sigma_a/\sigma_\perp$.





